Persistent current in metals with a large dephasing rate

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In a weakly disordered metal electron interactions are responsible for both decoherence of the quasi-particles as well as for quantum corrections to thermodynamic properties. We consider electrons which are interacting with two-level-systems. We show that the two-level-systems enhance the average equilibrium ("persistent") current in an ensemble of mesoscopic rings. The result supports the recent suggestion that two puzzles in mesoscopic physics may be related: The low temperature saturation of the dephasing time and the high persistent current in rings.

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Quantum interference effects play a crucial role in the low temperature properties of normal metals. Prominent examples are weak localization and the associated magnetoresistance. Recently it was suggested [1,2] that two of the unresolved problems in the physics of mesoscopic metals may have a common solution: The large value of the persistent current in mesoscopic rings and the low temperature saturation of the dephasing rate which is seen in magnetoresistance measurements.

The first problem is the large value of the persistent current in rings. Lévy et al [3] measured the nonlinear response to a magnetic field of an ensemble of 10^7 mesoscopic copper rings. The measured signal corresponds to a current $I \approx I_0 \sin(2\pi\phi/\phi_0)$ circulating in each ring. ϕ is the magnetic flux which penetrates each ring and $\phi_0 = h/e$ is the flux quantum. For temperatures in the mK regime the amplitude was $|I_0| \approx 0.3$ nA per ring, which is of the order of one elementary charge in the time τ_D an electron needs to diffuse around the ring, $|I_0| \approx 0.6e/\tau_D = 0.6eE_c/\hbar$. Here $E_c = \hbar/\tau_D = \hbar D/L^2$ is the Thouless energy, D is the electron diffusion constant, and L is the circumference of the ring. Similar results were reported in Refs. [1,4].

Theory, when neglecting electron interactions, predicts a current that is of the order $I \sim e\delta/\hbar$, where δ is the average distance of single particle levels at the Fermi energy [5-7]. With the parameters of the experiment [3], $\delta/k \approx$ 0.2 mK and $E_c/k \approx 25 \text{mK}$, the current obtained is about two orders of magnitude too small. Electron interactions first seemed to improve the situation [8]. For Coulomb interaction it was found that $I \sim e\mu^* E_c/\hbar$, where μ^* is a dimensionless number that characterizes the strength of the interaction in the Cooper channel. However estimates of μ^* gave a value which is an order of magnitude too small when comparing it with the experiment [8]. Surprisingly an enhancement of the current was also reported in presence of a moderate concentration of magnetic impurities [9], with $I \sim e(E_c/\hbar) \cdot \min(\hbar/\tau_s, E_c)/kT$, where $1/\tau_s$ is the spin-flip scattering rate. This can become larger than the current coming from the Coulomb interaction, however since the temperature dependence is different from the one observed this mechanism has not been considered as a possible explanation of the experiment in Ref. [3].

The second problem concerns the phase coherence of the electrons. Whereas it is expected that the dephasing rate goes to zero in the zero temperature limit [10] many experiments show a saturation at low temperature. Usually this saturation is attributed to the presence of magnetic impurities or to heating. However, recently a saturation of the dephasing time has been observed, also after excluding these possibilities [11,12]. Several attempts have been made to explain the low temperature saturation of the dephasing time [13–17]. It has been argued by Altshuler et al [14] that non-equilibrium electromagnetic noise can decohere the electrons without heating them. Originally, this non-equilibrium noise was suggested to be due to external radiation which couples into the samples. On the other hand dephasing could also occur due to internal noise. In this case a saturation of the dephasing time could also occur in equilibrium. Experimental evidence is in favor of an internal dephasing mechanism [11,12], however it is open if equilibrium or non-equilibrium processes dominate.

Recently Kravtsov and Altshuler [2] have extended earlier work [18] on the effect of a high frequency electromagnetic field in mesoscopic rings and have shown that non-equilibrium noise leads to a directed non-equilibrium current. They then suggested that both the "large" currents observed in [1,3,4] and the strong dephasing are related and non-equilibrium phenomena.

In this paper we demonstrate that also for the system in thermal equilibrium an enhanced persistent current is expected if there is an additional electron interaction which gives also rise to strong dephasing. For the particular model involving two-level-systems (TLS) we find (1) a diamagnetic current in the low magnetic field limit (2) a temperature dependence which is close to the experimentally observed one (3) an amplitude which depends on the concentration of TLS. In the following we first recall some of the theoretical concepts concerning the persistent currents. We then estimate the persistent cur-

rent coming from TLS and, finally, relate the persistent current amplitude and the dephasing rate.

The equilibrium current in a ring which is penetrated by a magnetic flux ϕ is calculated by taking the derivative of the thermodynamic potential, $I(\phi) = -\frac{\partial}{\partial \phi} \Omega(\mu, \phi)$. For simplicity we do not discuss the subtle questions concerning differences between the canonical $F(N,\phi)$ and the grand canonical thermodynamical potential $\Omega(\mu,\phi)$ [5–7]. In an ensemble of weakly disordered rings the disorder configuration will change from ring to ring, so in order to calculate the average persistent current of an ensemble of rings one has to average over disorder, $I(\phi) \rightarrow \langle I(\phi) \rangle_{\rm dis}$. For non-interacting electrons the grand canonical potential, and therefore the persistent current, depends only exponentially weak on the magnetic flux and one finds only a small persistent current [19].

The situation changes in presence of interactions. As an example take the Coulomb interaction as in Ref. [8] and consider the classical expression for the Coulomb energy,

$$H = \frac{1}{2} \int d\mathbf{r} d\mathbf{r}' v(\mathbf{r} - \mathbf{r}') \delta n(\mathbf{r}, \phi) \delta n(\mathbf{r}', \phi). \tag{1}$$

This quantity depends on the magnetic flux ϕ even on average since the fluctuations of the electron density are magnetic flux dependent and may be written as

$$\langle \delta n(\mathbf{r}, \phi) \delta n(\mathbf{r}', \phi) \rangle_{\text{dis}} = \sum_{m} A_m \cos(4\pi m\phi/\phi_0)$$
 (2)

with

$$A_{m} = \frac{4N(\epsilon_{F})}{\mathcal{V}} \frac{\sin^{2}(k_{F}|\mathbf{r} - \mathbf{r}'|)}{(k_{F}|\mathbf{r} - \mathbf{r}'|)^{2}} kT \sum_{\omega > 0} \sqrt{\frac{\omega}{E_{c}}} e^{-m\sqrt{\omega/E_{c}}},$$
(3)

compare Ref. [8,9]. Here $\omega=2\pi nkT$ are bosonic frequencies and $\mathcal V$ is the volume and $N(\epsilon_F)$ is the density of states of the Fermi level. As a technical remark we would mention that Eq.(3) is obtained by evaluating the diagram with one particle-particle propagator (cooperon). The harmonics of the persistent current $I(\phi)=\sum_m I_m \sin(4\pi\phi m/\phi_0)$ are finally found as $I_m=16\mu_0e/\pi m^2\tau_D$ with $\mu_0=N(\epsilon_F)\int \mathrm{d}\mathbf{r}v(\mathbf{r})\sin^2(k_Fr)/(k_Fr)^2$. Including the exchange energy reduces the current by a factor two, and higher orders in the interaction reduce the interaction amplitude, $\mu_0\to\mu^*\approx\mu_0/[1+\mu_0\ln(\epsilon_F\tau_D)]$.

When opening an additional interaction channel one will find an additional contribution to the persistent current. In Ref. [9] this has been demonstrated for magnetic impurities. Here we consider the interaction of conduction electrons with nonmagnetic impurities, which we assume to couple to the electron density. The Hamiltonian is of the form

$$\hat{H}_{\rm int} = \int \mathrm{d}x \hat{n}(\mathbf{x}) \hat{V}(\mathbf{x}). \tag{4}$$

The operator $\hat{V}(\mathbf{x})$ that is due to the impurities will be specified more explicitly below. To second order in this interaction one finds a correction to the free energy which is the sum of a Hartree and a Fock like term, $\delta\Omega = \delta\Omega_{\rm H} + \delta\Omega_{\rm F}$, which are given by $(\beta = 1/kT)$

$$\delta\Omega_{\rm H} = -\frac{1}{2} \int_0^\beta d\tau \int d\mathbf{x} \int d\mathbf{x}' \langle \hat{n}(\mathbf{x}) \rangle \langle \hat{n}(\mathbf{x}') \rangle$$

$$\times \left[\langle \hat{V}(\mathbf{x}, \tau) \hat{V}(\mathbf{x}', 0) \rangle - \langle \hat{V}(\mathbf{x}) \rangle \langle \hat{V}(\mathbf{x}') \rangle \right] \qquad (5)$$

$$\delta\Omega_{\rm F} = -\frac{1}{2} \sum_{s,s'} \int_0^\beta d\tau \int d\mathbf{x} \int d\mathbf{x}' \langle \Psi_s^{\dagger}(\mathbf{x}, \tau) \Psi_{s'}(\mathbf{x}', 0) \rangle$$

 $\times \langle \Psi_s(\mathbf{x}, \tau) \Psi_{s'}^{\dagger}(\mathbf{x}', 0) \rangle \langle \hat{V}(\mathbf{x}, \tau) \hat{V}(\mathbf{x}', 0) \rangle,$

(6)

where $\Psi_s^{\dagger}(\mathbf{x},\tau)$ and $\Psi_s(\mathbf{x},\tau)$ are operators for fermions with spin s and the brackets $\langle \ldots \rangle$ are the thermal average . If $\hat{V}(\mathbf{x})$ describes pure potential scattering, then $\hat{V}(\mathbf{x})$ is a c-number with the result that $\delta\Omega_{\rm H}=0$. $\delta\Omega_{\rm F}\neq0$ but does not depend on the magnetic flux which can be traced back to the fact that $\hat{V}(\mathbf{x},\tau) = \hat{V}(\mathbf{x})$ is static. This can become different if the impurity has an internal degree of freedom. Consider a TLS, realized by an impurity which sits in a double well potential with minima at \mathbf{r} and $\mathbf{r} + \mathbf{d}$ which are nearly degenerate in energy. We write the scattering potential as $\hat{V}(\mathbf{x}) = V[\hat{n}_A \delta(\mathbf{x} - \mathbf{r}) + \hat{n}_B \delta(\mathbf{x} - \mathbf{r} - \mathbf{d})].$ \hat{n}_A and \hat{n}_B are the number operators for the impurity in the relevant potential minimum. Since the impurity is in either of these minima $\hat{n}_A + \hat{n}_B = 1$. We further characterize the impurity by the asymmetry ϵ and a tunneling amplitude Δ between between the two minima, so the impurity Hamiltonian is

$$\hat{H}_{\rm imp} = \begin{pmatrix} \epsilon & \Delta \\ \Delta & -\epsilon \end{pmatrix}. \tag{7}$$

The Hartree energy (5), which is nonzero in this model, may be interpreted from the point of view of both the electrons and the impurities. From the electronic point of view the electron impurity interaction gives rise to an effective electron-electron interaction: Comparing Eqs.(1) and (5) one realizes that the Coulomb interaction is replaced by an effective interaction

$$v(\mathbf{x} - \mathbf{x}') \to -\int_0^\beta d\tau \left\{ \langle \hat{V}(\mathbf{x}, \tau) \hat{V}(\mathbf{x}', 0) \rangle - \langle \hat{V}(\mathbf{x}) \rangle \langle \hat{V}(\mathbf{x}') \rangle \right\}$$
(8)

due to the defects. From the impurity point of view the coupling to the conduction electrons changes the level asymmetry,

$$\begin{pmatrix} \epsilon & \Delta \\ \Delta & -\epsilon \end{pmatrix} \rightarrow \begin{pmatrix} \epsilon + V\langle \hat{n}(\mathbf{r}) \rangle & \Delta \\ \Delta & -\epsilon + V\langle \hat{n}(\mathbf{r} + \mathbf{d}) \rangle \end{pmatrix}, (9)$$

which then changes the free energy as given in Eq.(5) to second order in V.

We discuss the persistent current first in the most simple situation, where we neglect the tunnel splitting Δ . In this case $\hat{V}(\mathbf{x},\tau)$ is static so $\delta\Omega_{\mathrm{F}}$ remains independent of magnetic flux, as in the case of "normal" disorder. Here and below we will therefore concentrate on the Hartree energy. Using the relation $\hat{n}_A + \hat{n}_B = 1$ and averaging over "normal" disorder we can rewrite the Hartree energy as

$$\langle \delta \Omega_{\rm H} \rangle_{\rm dis} = -|V|^2 \langle \delta n^2(\mathbf{r}, \phi) \rangle_{\rm dis} \left(1 - \frac{\sin^2(k_F d)}{(k_F d)^2} \right)$$
$$\times \int_0^\beta d\tau \left\{ \langle \hat{n}_A(\tau) \hat{n}_A(0) \rangle - \langle \hat{n}_A \rangle \langle \hat{n}_A \rangle \right\}. \quad (10)$$

If the TLS asymmetry is large, $|\epsilon| > kT$, then $\langle \hat{n}_A(\tau)\hat{n}_A(0)\rangle - \langle \hat{n}_A\rangle \langle \hat{n}_A\rangle = 0$ and therefore $\langle \delta\Omega_{\rm H}\rangle_{\rm dis} = 0$. For a TLS with a small asymmetry, $|\epsilon| < kT$ one finds $\langle \hat{n}_A(\tau)\hat{n}_A(0)\rangle - \langle \hat{n}_A\rangle \langle \hat{n}_A\rangle = 1/4$ so that $\langle \delta\Omega_{\rm H}\rangle_{\rm dis} \neq 0$ and a persistent current results. From the integration over τ it follows that the current coming from a single defect is proportional to the inverse temperature, in full analogy to the persistent current from a magnetic impurity [9]. For the system with a finite density of TLS the asymmetry ϵ will not be a constant, instead there will be a distribution of asymmetries. Using eq.(2) and below we determine the persistent current as

$$I \approx -\frac{8}{\pi} \frac{c_{\text{act}} N(\epsilon_F) V^2 F}{kT} \frac{e}{\tau_D},\tag{11}$$

where $F = 1 - \sin^2(k_F d)/(k_F d)^2$ and c_{act} is the concentration of TLS with $\epsilon < kT$ and therefore is active in producing a persistent current. Assuming a flat distribution of asymmetries between zero and $\epsilon_{\text{max}} > kT$, the concentration of active TLS is proportional to the temperature, $c_{\rm act} = ckT/\epsilon_{\rm max}$, which then cancels the inverse temperature dependence of the persistent current of a single defect. The current is diamagnetic in contrast to the paramagnetic current from the repulsive Coulomb interaction. The amplitude of the current is of the diffusive scale, $I \sim e/\tau_D$, as for the Coulomb interaction. The dimensionless prefactor μ^* is replaced by the factor $\mu_{\rm TLS} = -cFN(\epsilon_F)V^2/\epsilon_{\rm max}$. which should be of order one if this mechanism is relevant for the currents observed in Ref. [3]. Assuming an atomic scattering cross section of the TLS and the factor $F \sim 1$ this requires a density of states of TLS that is comparable to the density of states of the metallic host and therefore of the order 10¹⁸/Kcm³. At 100mK this corresponds to a concentration of active two-level-systems of about 2ppm which is not a small number but, in principle not impossible [20]. For the assumed distribution of asymmetries the temperature dependence of the persistent current is only due to the temperature dependence of the local density fluctuations, see eq.(3), and is therefore identical to the temperature dependence of the persistent current from the Coulomb interaction. The latter has been shown [8]

to agree well with the experiment in Ref. [3]. Finally it is important to discuss spin-orbit scattering, since in the gold or copper rings in the experiments the spin-orbit rate is large. Following Refs. [8,9] we find that spin-orbit scattering reduces the persistent current due to the mechanism discussed here by a factor four, but the sign remains diamagnetic.

Let us now allow a finite tunnel splitting Δ , i.e. spontaneous transitions of the impurity between the two minima. The correlation function that is relevant for the persistent current, i.e. the impurity susceptibility, is given by

$$\int_{0}^{\beta} d\tau \langle \hat{n}_{A}(\tau) \hat{n}_{A}(0) \rangle - \langle \hat{n}_{A} \rangle \langle \hat{n}_{A} \rangle = \begin{cases} \frac{1}{4} \frac{1}{kT} \\ \frac{1}{4} \frac{\Delta^{2}}{\epsilon^{2} + \Delta^{2}} \frac{1}{\sqrt{\epsilon^{2} + \Delta^{2}}} \end{cases} ,$$
(12)

in the two limits where $\epsilon^2 + \Delta^2 < (kT)^2$ and $\epsilon^2 + \Delta^2 > (kT)^2$. Whereas for static defects with $\Delta = 0$ the correlation function is non-zero only in the high temperature limit, $kT > \epsilon$, the correlation function for dynamic defects is non-zero even in the zero temperature limit, so these defects contribute to the persistent current even for $T \to 0$. We calculate the persistent current under the assumption [21] of a flat distribution of ϵ between zero and $\epsilon_{\rm max}$ and a distribution of Δ that is proportional to $1/\Delta$ between $\Delta_{\rm min}$ and $\Delta_{\rm max}$. We then find $I \sim -(e/\tau_D)F\hbar/(\tau_{\rm TLS}\epsilon_{\rm max})$ as before when we neglected the tunnel splitting. $\hbar/\tau_{\rm TLS} \sim cN(\epsilon_F)V^2$ is the electron scattering rate off the TLS.

Finally we discuss the relation of the persistent current and dephasing. In Ref. [15] it has been demonstrated that TLS lead to dephasing with a rate that is temperature independent in a certain range of temperature. Both the persistent current amplitude and the dephasing rate are hard to estimate for a given material since they depend on the concentration of TLS and the distribution of ϵ and Δ . It is therefore of interest to relate the two quantities, in order to reduce the number of unknown parameters. Notice that in order to have dephasing there must be real transitions between two impurity states, and one finds that the defects with $kT > \sqrt{\epsilon^2 + \Delta^2} > \hbar/\tau_\phi$ are most effective for dephasing. On the other hand all defects with $kT > \sqrt{\epsilon^2 + \Delta^2}$ and even some with $kT < \sqrt{\epsilon^2 + \Delta^2}$ contribute to the persistent current, see Eq.(12). We cannot therefore give a general relation between dephasing rate and persistent current amplitude. We can, however, as shown below, give such a relation for our special choice of the distribution of ϵ and Δ . The dephasing rate has been estimated as [15]

$$\frac{1}{\tau_{\phi}} \sim \begin{cases} \Delta_{\max} F / (\epsilon_{\max} \tau_{\text{TLS}} \lambda) & \text{if } \hbar / \tau_{\phi} < \Delta_{\max} < kT \\ \Delta_{\max} (F / \hbar \lambda \epsilon_{\max} \tau_{\text{TLS}})^{1/2} & \text{if } \Delta_{\max} < \hbar / \tau_{\phi} < kT \end{cases}$$
(13)

with $\lambda = \ln(\Delta_{\text{max}}/\Delta_{\text{min}})$. The persistent current amplitude, $I \sim \mu_{\text{TLS}}(e/\tau_D)$ with $|\mu_{\text{TLS}}| \sim F\hbar/(\epsilon_{\text{max}}\tau_{\text{TLS}})$, is therefore of the order

$$|\mu_{\rm TLS}| \sim \begin{cases} \lambda(\hbar/\tau_{\phi})/\Delta_{\rm max} \\ \lambda(\hbar/\tau_{\phi})^2/\Delta_{\rm max}^2 \end{cases}$$
 (14)

in the two limits considered. For example for the gold sample of Ref. [1] $\hbar/\tau_{\phi} \sim 2 \, \mathrm{mK}$ below 500mK. If the constant dephasing rate is from the mechanism we consider, then the lowest measured temperature ($\sim 40 \, \mathrm{mK}$) is an upper limit for Δ_{max} , and leads to the estimate $|\mu_{\mathrm{TLS}}| > \lambda/20$.

The dephasing rate for low temperature, $\hbar/\tau_{\phi} < kT < \Delta_{\rm max}$, is proportional to T [15] and given by $1/\tau_{\phi} \sim FkT/(\epsilon_{\rm max}\tau_{\rm TLS}\lambda)$. Here one finds $|\mu_{\rm TLS}| \sim \lambda(\hbar/\tau_{\phi})/kT$, which depends only on one unknown parameter, λ . A dephasing rate which is linear in T has been observed in various three-dimensional and two-dimensional samples [22]. The values which were reported for τ_{ϕ} at 10K are around $\tau_{\phi} \sim 10^{-12} {\rm sec} - 5 \cdot 10^{-10} {\rm sec}$, which corresponds to $(\hbar/\tau_{\phi})/kT \sim 2 \cdot 10^{-2} - 1$. Also from these considerations it seems rather reasonable that the parameter $|\mu_{\rm TLS}|$ can reach values of order one.

In this paper we estimate the persistent current linear in the concentration of TLS and we neglect Kondo correlations. Kondo physics has been suggested as a possible solution of the dephasing problem in Ref. [16]. The persistent current, of course, will be modified by Kondo correlations, however it is beyond the scope of this paper to estimate this quantitatively. We also do not attempt to give an exhaustive discussion of the limit of high concentration of impurities. However it is clear that our theory will overestimate the current when the phase coherence time τ_{ϕ} becomes of the order of, or shorter than, the diffusive time τ_D . The related problem for the persistent current coming from magnetic impurities has been discussed in Ref. [9].

In summary we demonstrated that the interaction of conduction electrons with impurities induces a persistent current. Under reasonable assumptions we find a temperature dependence that is set by the diffusive scale. The most crucial point however is the current amplitude here given by $I \sim -|\mu_{\rm TLS}|e/\tau_D$. The dimensionless parameter μ_{TLS} depends on the concentration of the TLS, so a reliable estimate of the current amplitude is difficult. Experimentally the interrelationship of dephasing and persistent current may be checked by measuring the persistent current for different materials. For silver, where no saturation of the dephasing time has been observed [12], we expect a smaller persistent current than in gold or copper where the dephasing time saturates at low temperature. The sign of the current may help to decide if non-equilibrium fluctuation suggested in Ref. [2] or the equilibrium electron-impurity interactions studied here dominate the current: For a system with strong spin-orbit interactions Ref. [2] predicts a paramagnetic current, whereas we found a diamagnetic current.

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